Plutonium: An anthropogenic element under nuclear forensic investigations

Maria Wallenius

Institute for Transuranium Elements (ITU)
Karlsruhe, Germany

http://itu.jrc.cec.eu.int

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Plutonium is an anthropogenic, i.e. man-made element.

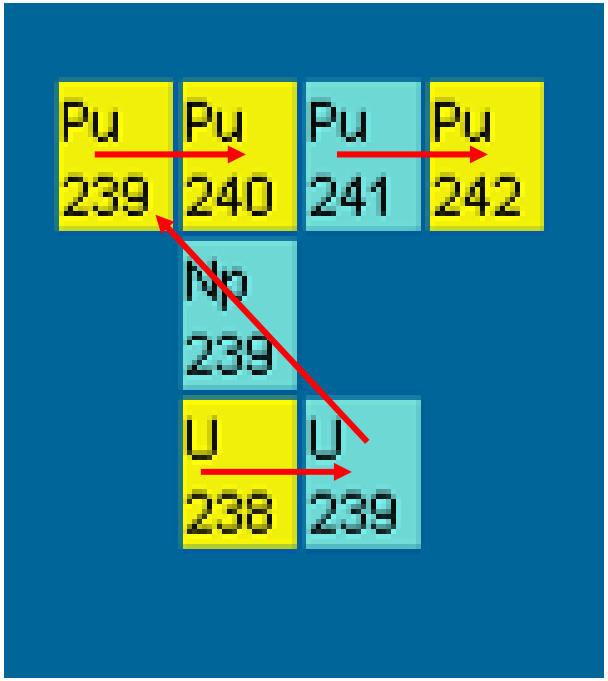
At the end of 1940 Pu-238 was first time synthesized:

U-238 (d,2n) Np-238 $\xrightarrow{\beta-}$ Pu-238

In nuclear reactor (~1% Pu in spent fuel):

U-238 + n $\rightarrow$ U-239 $\xrightarrow{2\ \beta-}$ **Pu-239** + n $\rightarrow$ **Pu-240** + n $\rightarrow$ **Pu-241** + n $\rightarrow$ **Pu-242**

U-235 + 2n $\xrightarrow{\beta-}$ U-237 $\xrightarrow{\beta-}$ Np-237 + n $\rightarrow$ Np-238 $\rightarrow$ **Pu-238**



Nuclear forensics

Main questions to be answered:

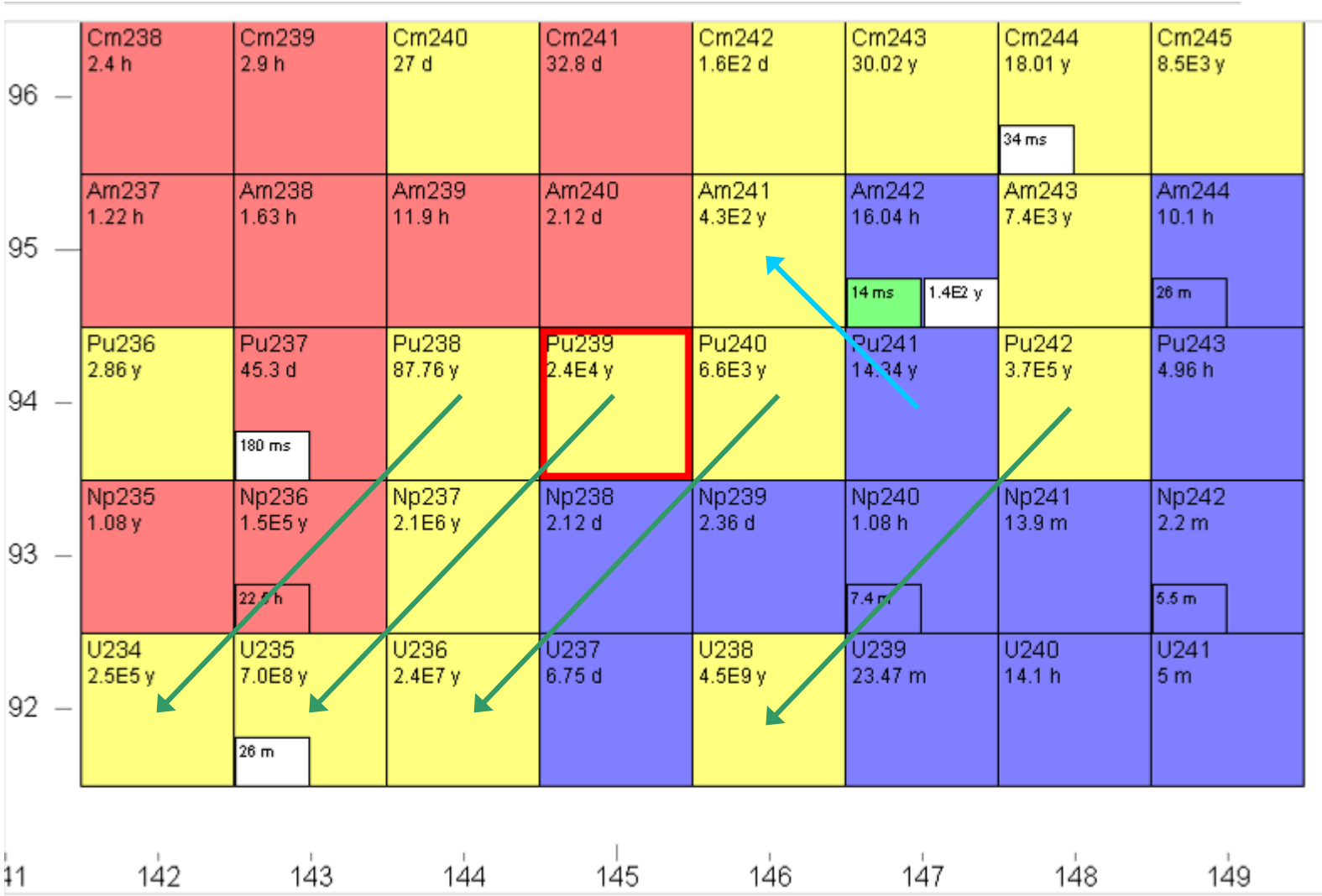
1. **Intended use** of the material
   (nuclear weapons, fuel, scrap,…)

2. **Origin**
   (place of production, process, place of diversion, last legal owner,…)

Answers:

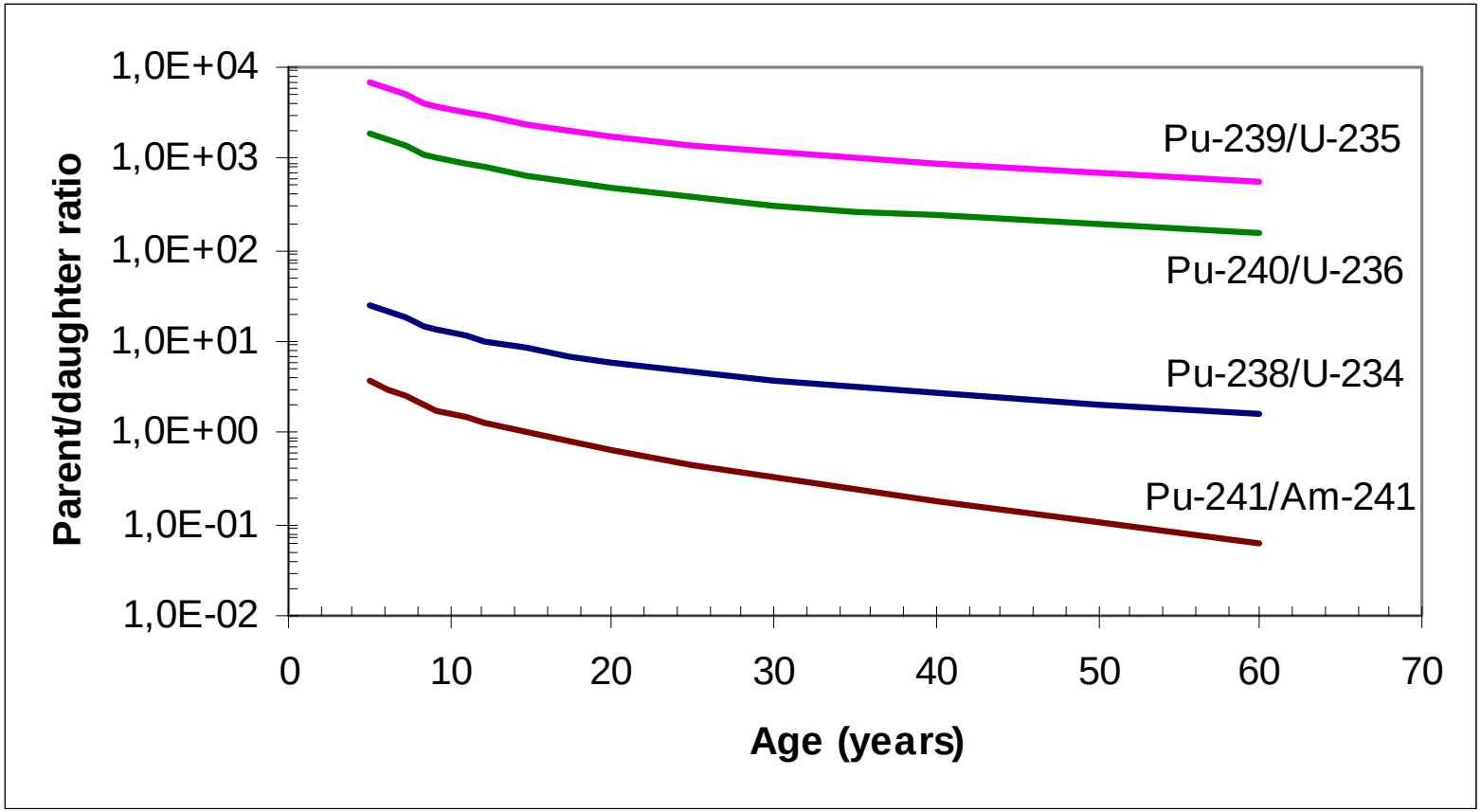
- isotopics
- Pu-content
- age
- impurities
- microstructure
- …

Plutonium decay



Age determination

Age = the time elapsed since the last chemical processing (e.g. production, reprocessing, purification)



Age determination - limitations

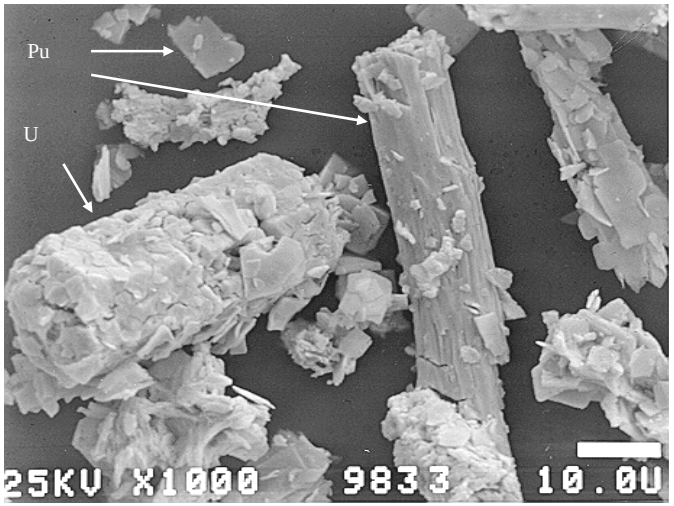
- Incomplete Pu/U/Am separation when reprocessing

  ⇒ U and Am remainings

  ⇒ inconsistent results

Sample	$^{241}Pu/^{241}Am^{*}$	$^{238}Pu/^{234}U$	$^{239}Pu/^{235}U$	$^{240}Pu/^{236}U$
RR	6.1	**2.35** ± 0.10	**2.66** ± 0.26	**2.29** ± 0.05
F19	20.6	**1.18** ± 0.07	**51.9** ± 3.8	**19.9** ± 0.3

Age by SIMS in February 2000. * By gamma spectrometry.

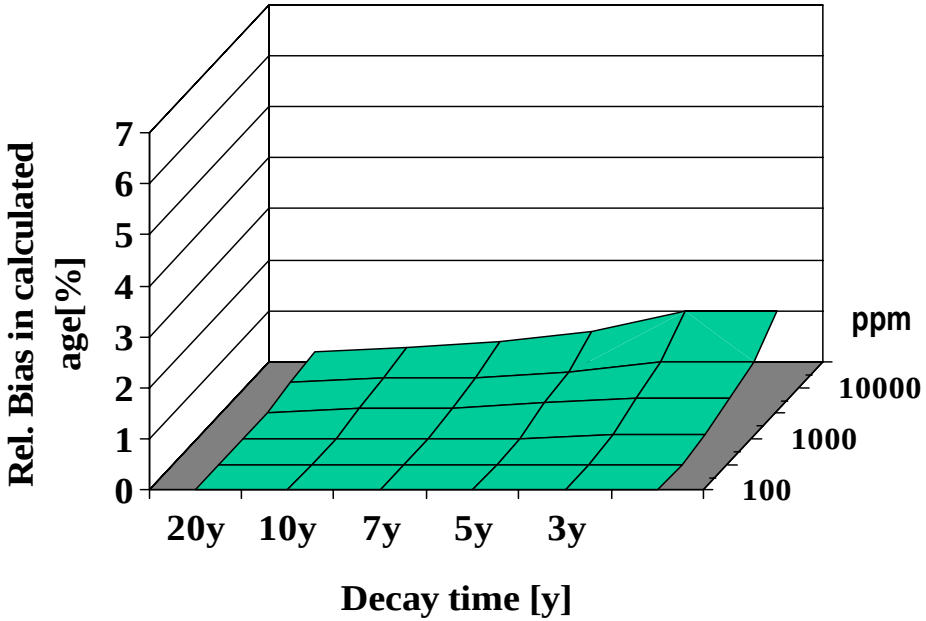


- MOX

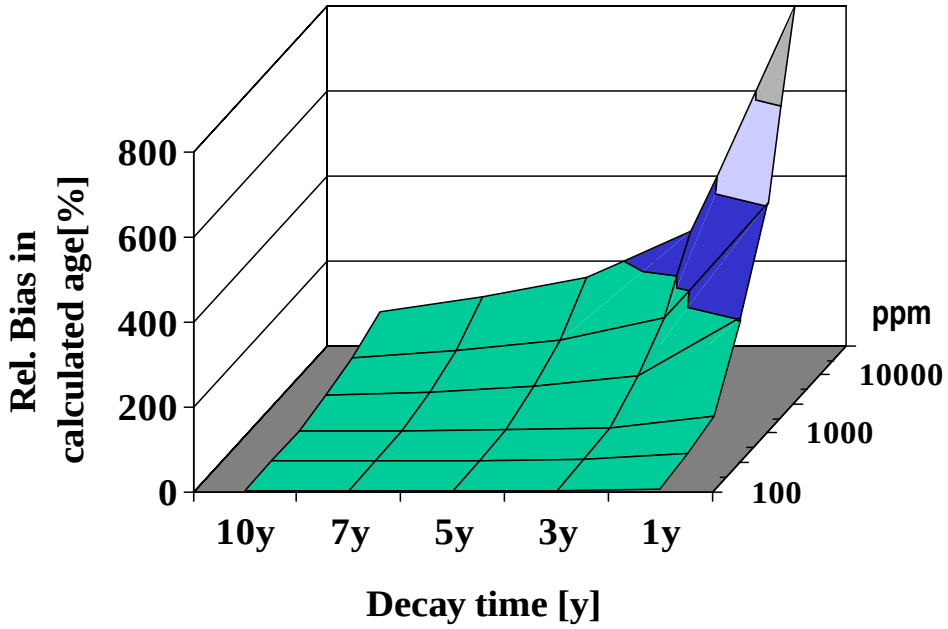
  ⇒ U interference

Effect of the residual U on the age

^{238}Pu / ^{234}U
reactor grade



^{238}Pu / ^{234}U
weapons grade



Plutonium composition

Weapons grade:

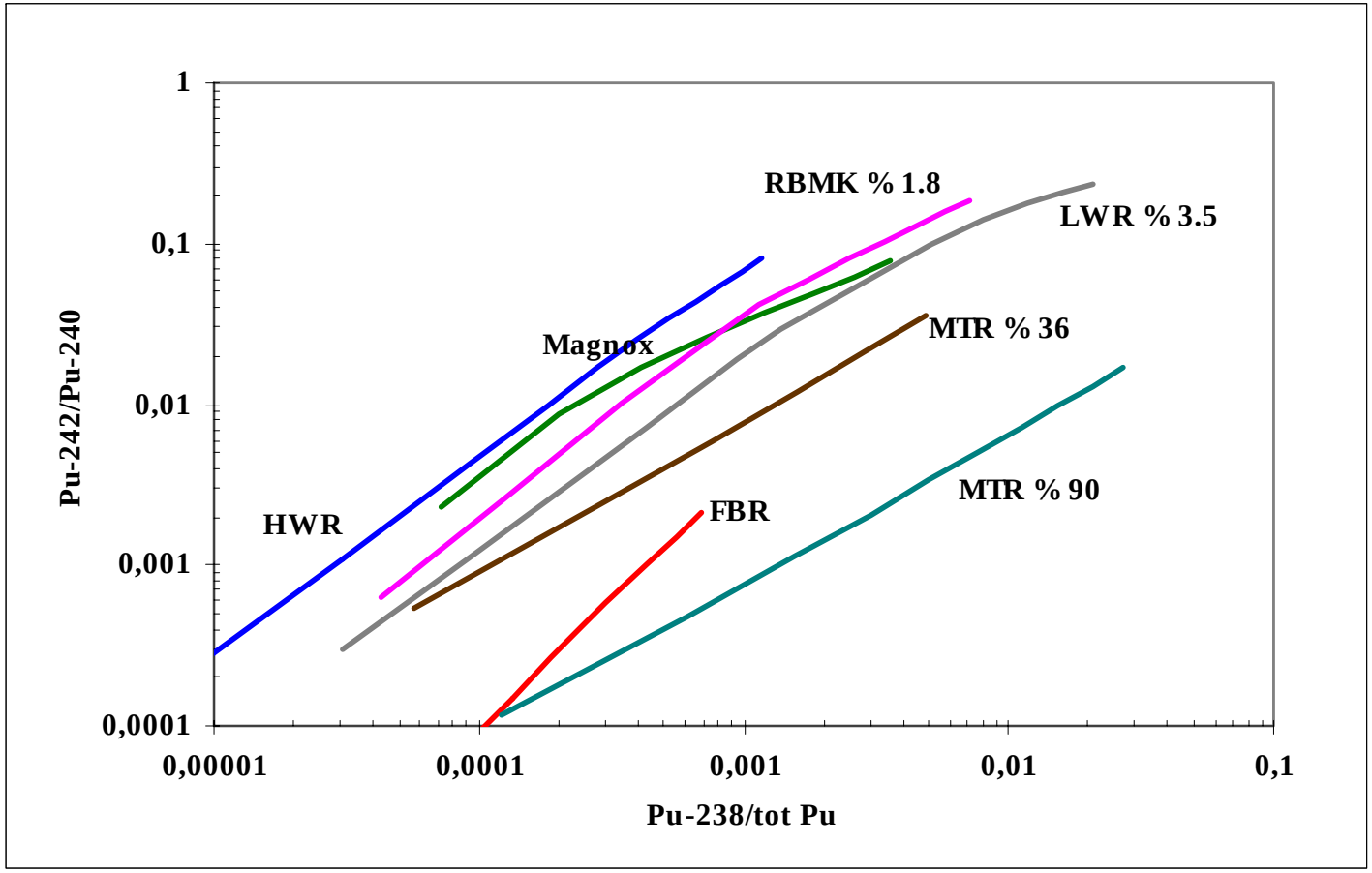
- Pu-240 < 7 %
- 240/239 < 0.07

Reactor grade:

- Pu-239 = 45-60%
- Pu-240 = 20-35 %
- 240/239 = 0.3-0.8

The isotopic composition of the reactor produced plutonium is depending on the initial U-235 enrichment, neutron spectrum (soft >< hard), flux and irradiation time.

Reactor type determination

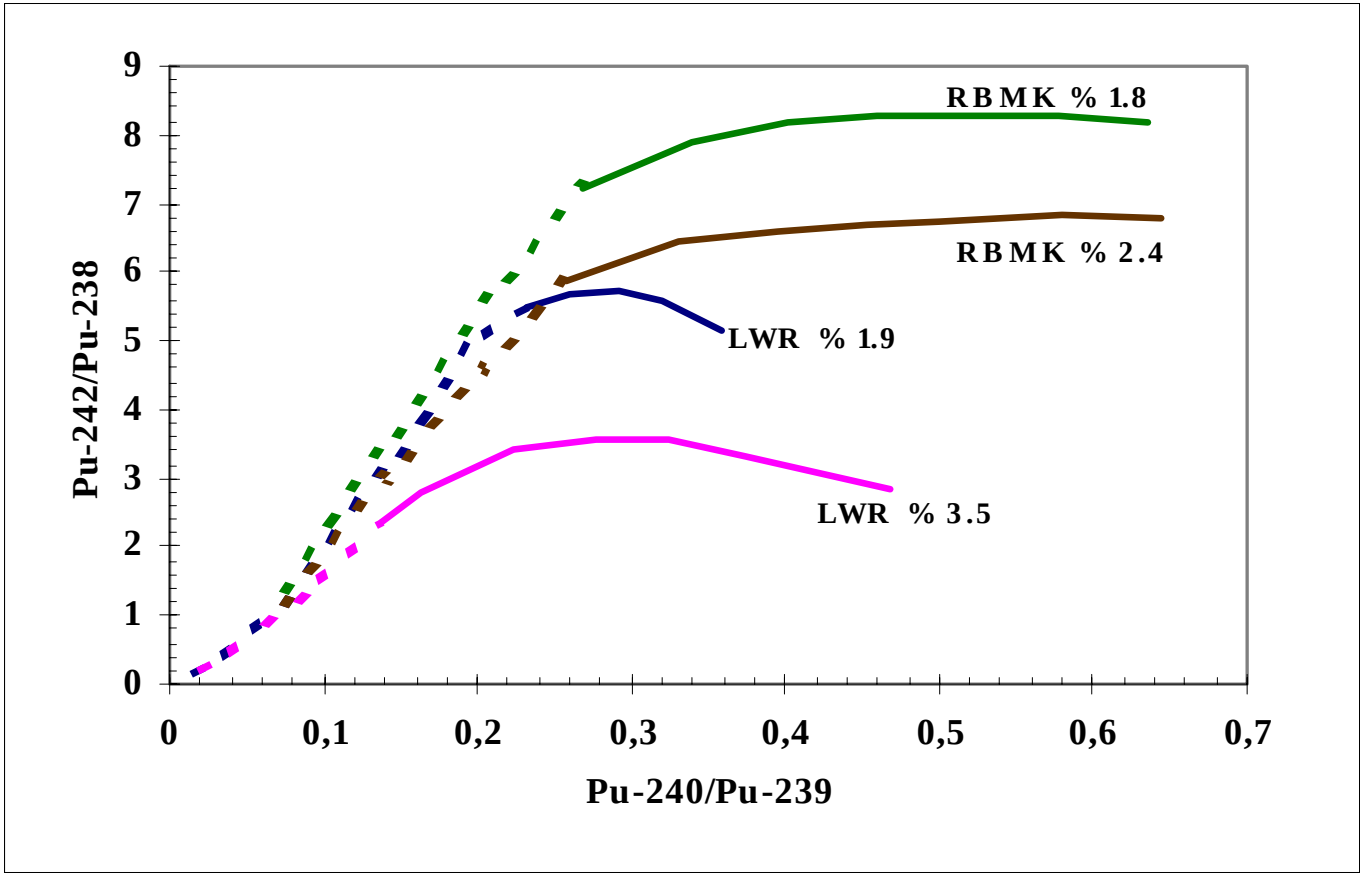


Softer the neutron spectrum – higher the $^{242}Pu/^{240}Pu$ ratio

Higher the initial ^{235}U enrichment – higher the ^{238}Pu abundance

Reactor type determination

Difference between light-water and graphite-moderated reactors.



Case studies

Find 13- Konstanz

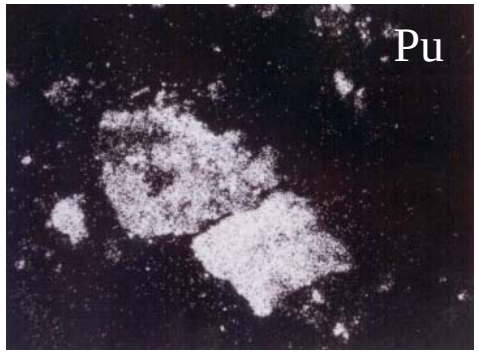
56 g of powder containing Pu

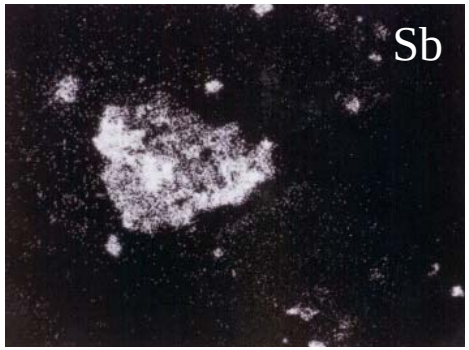
Isotope/ element	w-%
^{238}Pu	0.067
^{239}Pu	99.751
^{240}Pu	0.181
Mercury	61
Antimony	11
Oxygen	6
Plutonium	10
Gallium	0.15 (4.6 a-%)
Iodine	1.6

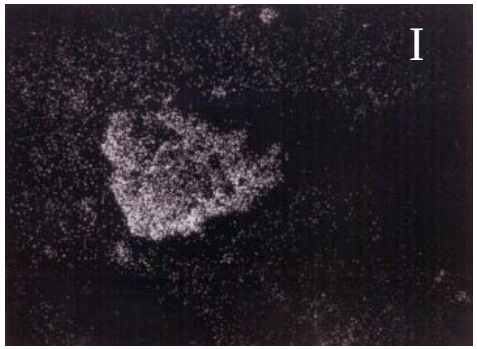
$Hg_2Sb_2O_7$ = “Red mercury” !

Find 13- Konstanz

Pu particle and powder agglomerate - electron probe







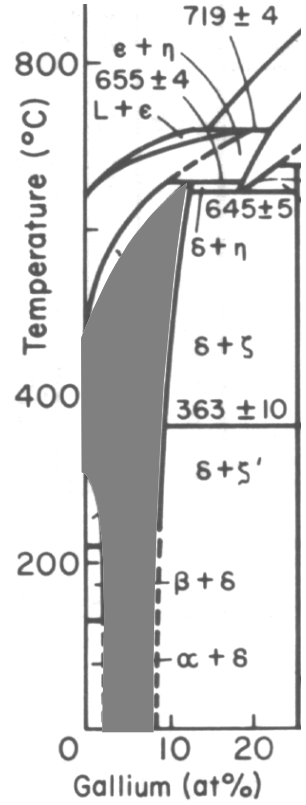
Find 13- Konstanz

“Red mercury”:

- Anti-Radar-Shielding for aircraft’s
- self-guided warheads
- nuclear bombs smaller and more efficient
- density 23 g/ccm

Plutonium:

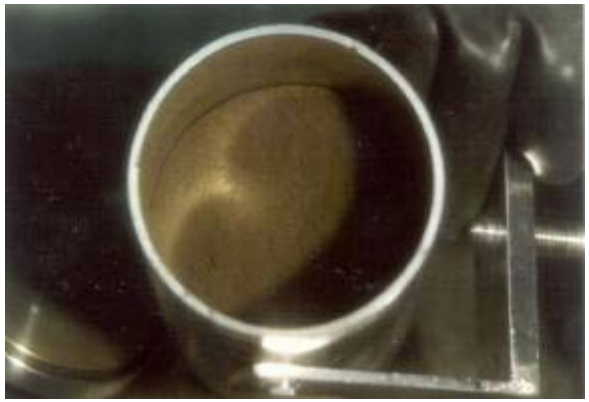
- weapons-grade Pu
- Ga is used to stabilise the δ- phase of Pu



Compound with these characteristics has never been found !!

Example of the proliferation of classified knowledge !

Find 19 - Munich



Mixed plutonium and uranium oxides:

363 g Pu - 121 g U

Pu	238	239	240	241	242
w-%	0.17	87.58	10.78	0.81	0.66

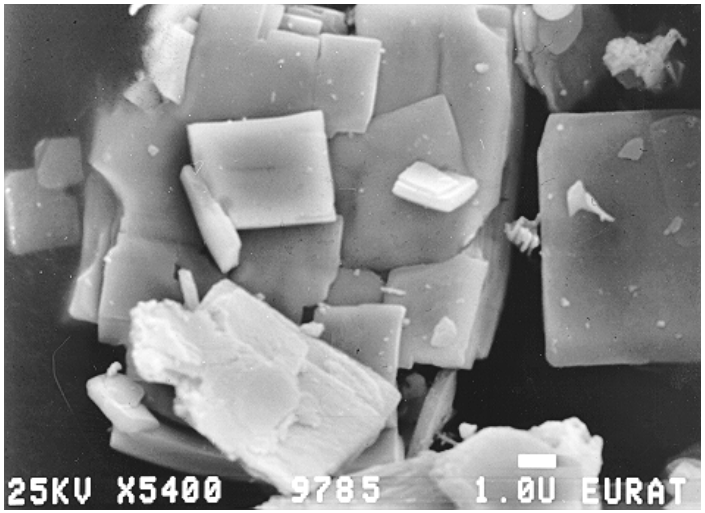
U	234	235	236	238
w-%	0.020	1.60	0.048	98.35

201 g of Li-metal enriched to 89.4 % in ^{6}Li.

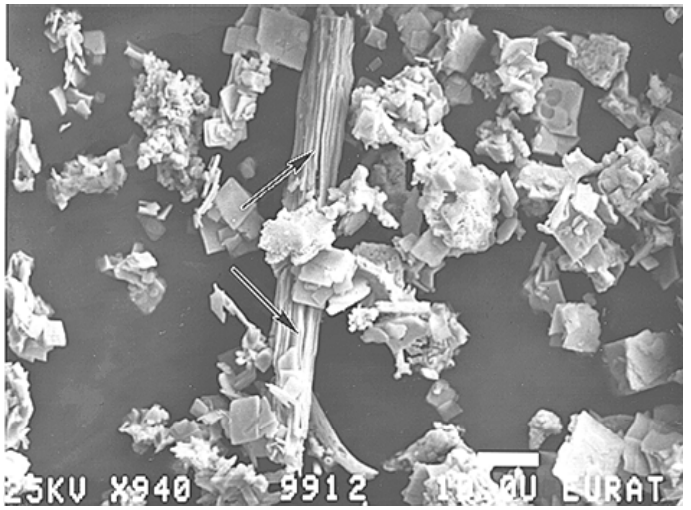
Find 19 - Munich

Powder consists of two different Pu particle types:

1) platelets of PuO_2



2) rod-shaped PuO_2

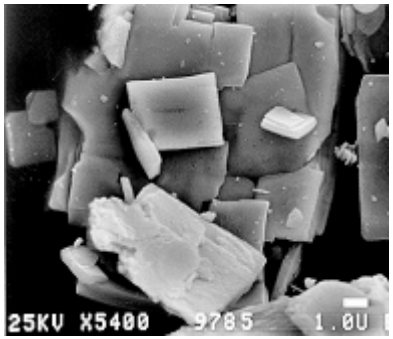


	Bulk	Particle 1	Particle 2
$^{240}Pu/^{239}Pu$	0.1226	0.1159	0.1245
Age from the $^{240}Pu/^{236}U$*	(20.6)**	21.5	19.9

*Age by SIMS in February 2000. ** By gamma spectrometry from the Am-241 build-up.

Find 19 - Munich

Microstructural fingerprint



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SEM

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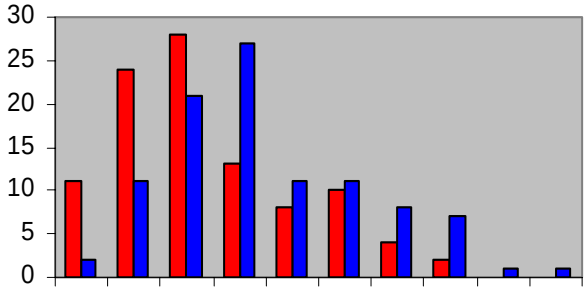
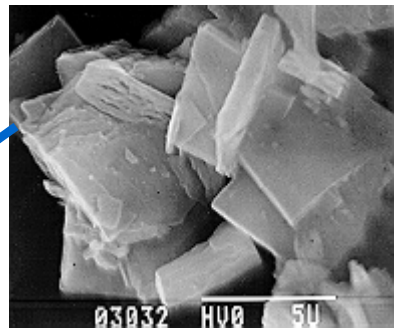
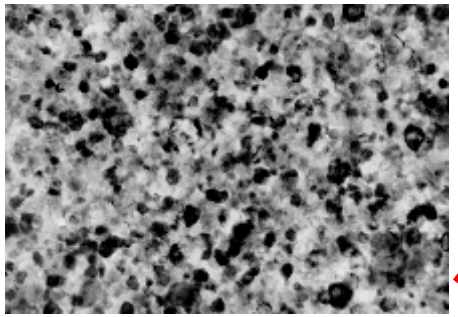




Reference SM3-1



TEM

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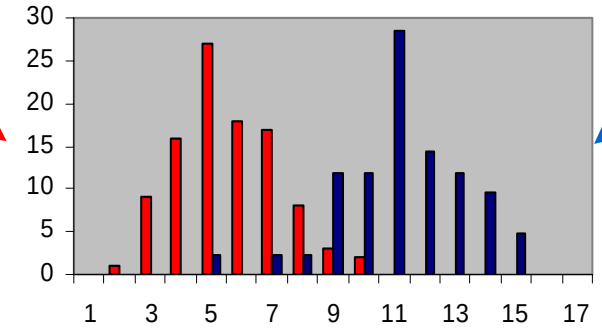
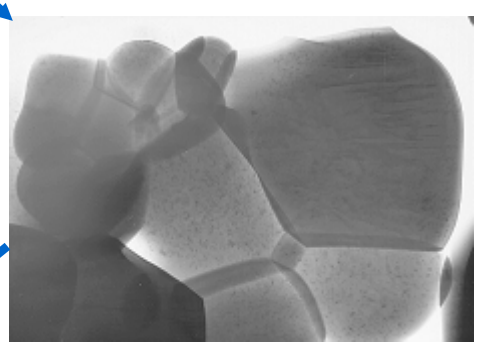




Find 19 - Munich

Plutonium

- close to weapons-grade
- not from commercial reprocessing or from military production
- mixture of residues of different types of spent fuels

Lithium-6

- energetic tritons via $^{6}Li(n,\alpha)^{3}T$
- tritium can initiate DT nuclear fusion in a thermonuclear weapon

May not be a coincidence to find such materials together !!

Find 24 - Karlsruhe

WAK – Reprocessing plant (decommissioned)

Contamination of an employee was detected in a routine urine control.

Find 24 - Karlsruhe

“Players”:

Mr. M. – The thief

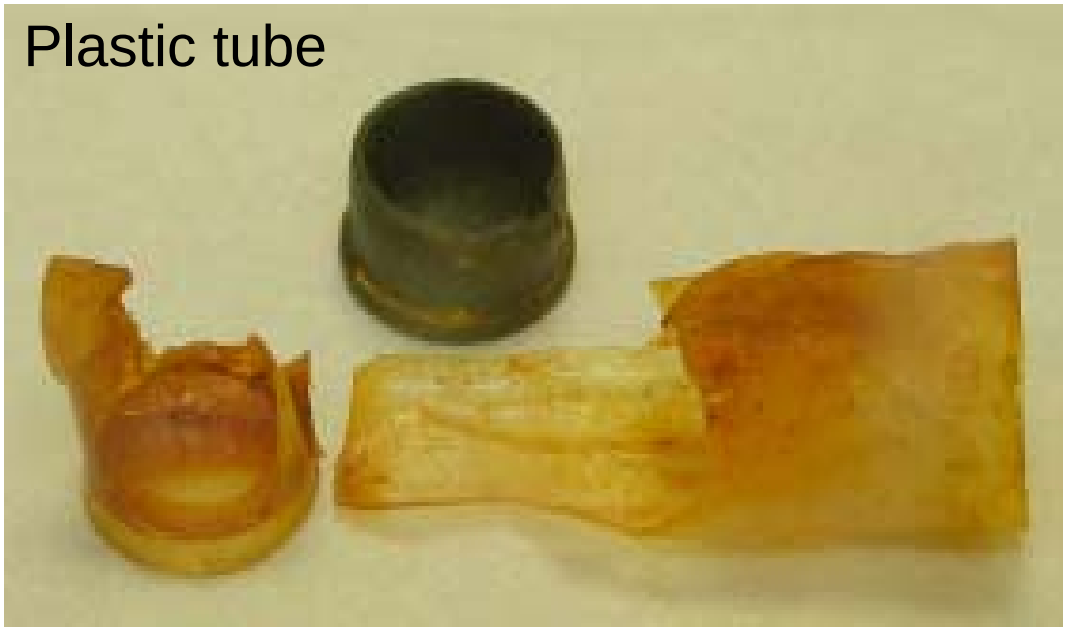
Mrs. E. - Girl friend of Mr. M.

Ms. A. E. - Daughter of Ms. E.

Find 24 - Karlsruhe

Stolen items:

Plastic tube



Swipe cloth

Total of a few milligrams of radioactive material including Pu, U, ^{241}Am, $^{134+137}$Cs, ^{154}Eu and ^{125}Sb.

Find 24 - Karlsruhe

Estimated (ICRP Model) activity uptake (in kBq):

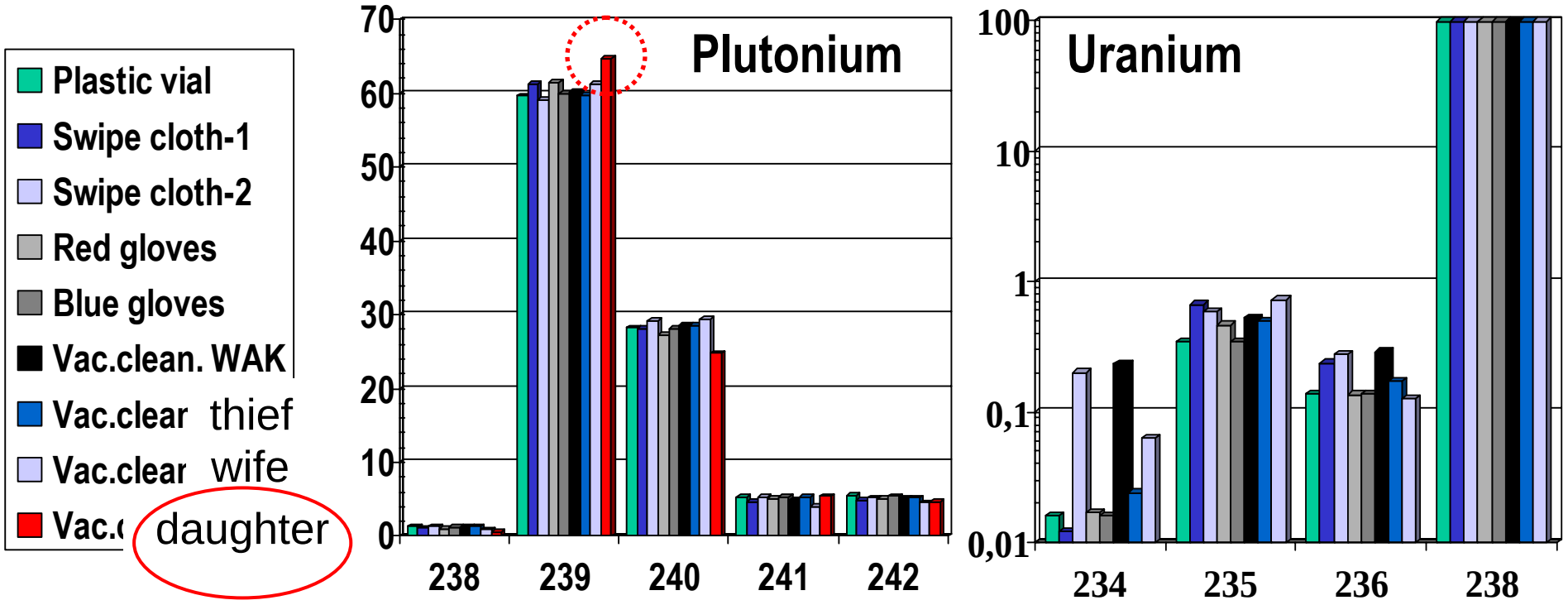
Person	Way of uptake	Cs-137	Am-241	Pu-241
Mr. M.	Inhalation		4	16
	Ingestion		-	-
Mrs. E.	Inhalation		28	1110
	Ingestion	118	3450	13800
Ms. A.E.	Inhalation		0,4	
	Ingestion	3	69	

Find 24 - Karlsruhe

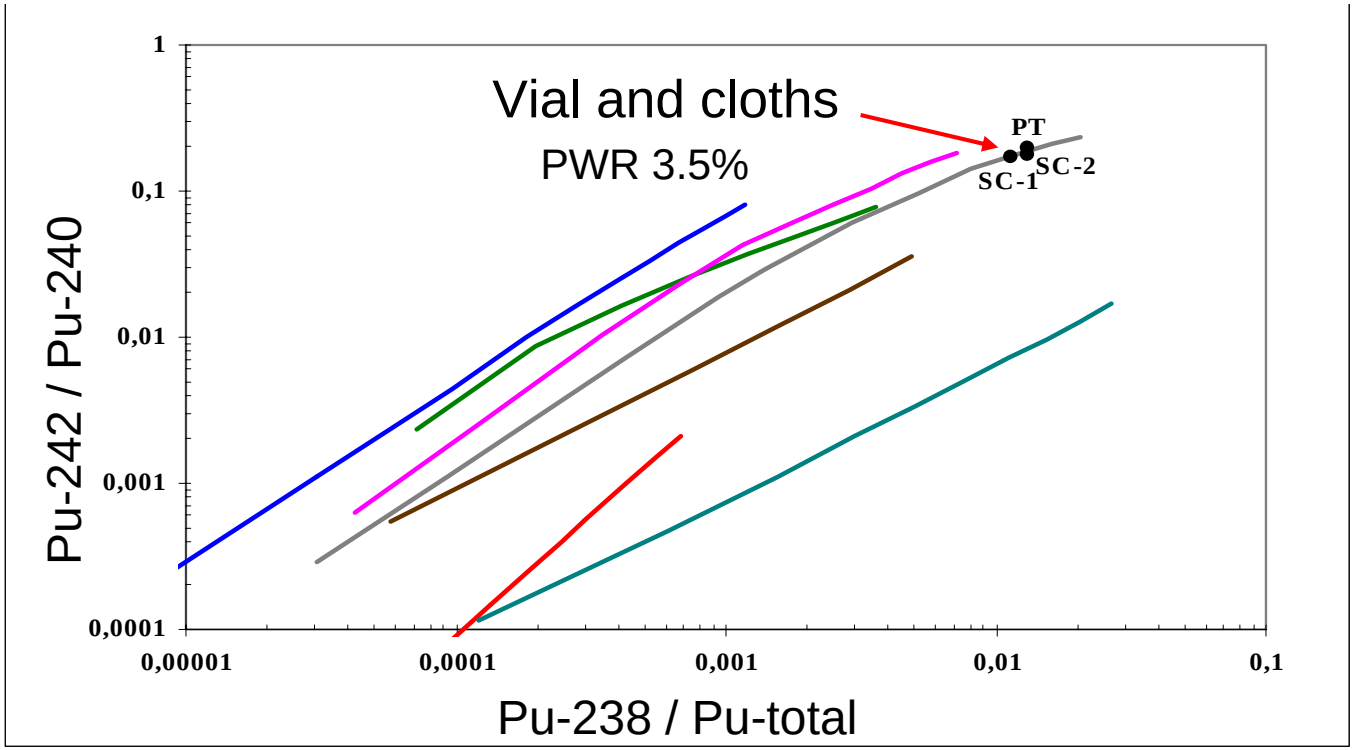
Can the contaminations and incorporations be explained by the two sources, vial + cloth ?

- Analyse samples from different locations
  - three apartments (thief, girl friend, daughter's boyfriend)
  - facility
- Compare
  - samples
  - external information (which fuel reprocessed, WAK, …)

Isotopic compositions:



Find 24 - Karlsruhe



– Pu composition matched with the reprocessed LWR fuel

– The age of the Pu was 12-14 years, which was the time of the last reprocessing campaign

Find 24 - Karlsruhe

- Vacuum cleaner bag from “the daughter” showed significantly different Pu isotopic composition

  ⇒ Pu-240/Pu-239 = 0.38 rather than 0.47

  ⇒ mixture of reactor Pu (~ 90%) and fallout Pu (~10%)

- The plastic vial can be excluded as source of incorporation of the wife (^{137}Cs content too low)
  Swipe cloth:
  Varying elemental ratios can be explained by different solubilities (Cs-137 dissolves well in water, Pu and Am not)

- Decontamination of two apartments ~ 2 M€

Exercises

Exercises

1. Which parent/daughter and parent/grand-daughter ratios can be used to determine the age of a 10 mg Pu-sample with the following composition (wt-%):

   Pu-238: 2 %
   Pu-239: 57 %
   Pu-240: 25 %
   Pu-241: 10 %
   Pu-242: 6 %

   Remarks: Method of the choice is ICP-MS without chemical separation with detection range of 10^8 (i.e. the parent/daughter ratio cannot be larger than 10^8).

2. In which of the following reactors was the Pu from previous example most likely produced ?

- BWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- PWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- BWR, 1.9 % U-235, 22 Burn-up, 8 years decay

Remember: Pu-240/Pu-239 = 0.44
Pu-241/Pu-239 = 0.18